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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/042,181	01/11/2002	Nestor A. Bojarczuk JR.	YOR9-2000-0644	5965

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EXAMINER	
SMITH, BRADLEY	

ART UNIT	PAPER NUMBER
2824	

DATE MAILED: 03/24/2004

Please find below and/or attached an Office communication concerning this application or proceeding.

Office Action Summary	Application No.	Applicant(s)	
	10/042,181	BOJARCZUK ET AL.	
	Examiner	Art Unit	
	Bradley K Smith	2824	AW

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on *31 December 2003*.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) *1-14,24-31,33,34,36,37 and 39* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *1-14,24-31,33,34,36,37 and 39* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☒ The drawing(s) filed on *29 March 2003* is/are: a)☒ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

a)☐ All b)☐ Some * c)☐ None of:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☐ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application (PTO-152)
6) ☒ Other: *Search notes*.

DETAILED ACTION

Claim Rejections - 35 USC § 102

1. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for a patent.

2. Claims 24 and 25 are rejected under 35 U.S.C. 102(a) as being anticipated by Chon et al. "Fatigue free samarium-modified bismuth titanate film capacitors having a large spontaneous polarizations". Chon et al. disclose a rare earth based memory element based on a hysterisis and current voltage characteristics (Figure 2). Furthermore Chon et al. disclose the a substrate a metal oxide layer and a conductive layer on the metal oxide layer.

Claim Rejections - 35 USC § 103

2. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

2. Claims 1-3, 5-8, 10 and 13 are rejected under 35 U.S.C. 103(a) as being unpatentable over Haukka et al. (US Pregrant Publication 2002/01 15252) Haukka et al. disclose a gate dielectric with a substrate comprising of a semiconductor material , a metal oxide layer comprising an electrically insulating rare earth metal oxide disposed on the surface of the substrate , a conductive material on metal oxide layer and the

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paragraph 0008

conductive material acting as a gate electrode (figure 3). However, with respect to claim 1, Huakka fails to disclose that a second electrode is attached to the substrate, and with respect to claim 10, Haukka et al. fails to disclose a second and third electrode attached to the source and drain region respectively. But the examiner takes official notice that since Haukka et al. disclose that this invention is for a transistor, and transistors can be memory elements (i.e. RS Latch). It would be obvious to one of ordinary skill in the art at the time the invention was made, to attach electrodes to the substrate more specifically the source and drain regions, because a solid state transistor by definition has to have a electrodes attached to the source and drain in order to function. Furthermore with respect to claims 2 , 3, 5 and 13, Haukka et al. disclose the dielectric layer comprises a mixture of lanthanum oxide and aluminum oxide. With respect to claims 6 and 7, Haukka et al. disclose the metal oxide layer has a thickness of 30-90 angstroms. With respect to claim 8, Haukka et al. disclose the conductive material comprises polysilicon.

3. Claims 4, 9, 14, 28 and 29 are rejected under 35 U.S.C. 103(a) as being unpatentable over Haukka et al. (US Pregrant Publication 2002/01 15252). Haukka et al. disclose the claimed invention except for the conductive material is aluminum. It would have been obvious to one having ordinary skill in the art at the time the invention was made to use aluminum as the conductive material, since it has been held to be within the general skill of a worker in the art to select a known material on the basis of its suitability for the intended use as a matter of obvious design choice. In re Leshin,

125 USPQ 416. Furthermore aluminum has a is a good conductor and the use of aluminum as a conductive material is well known in the art.

4. Claims 11, 12, and 30 are rejected under 35 U.S.C. 103(a) as being unpatentable over Haukka et al. (US Pregrant Publication 2002/01 15252). Haukka et al. disclose the claimed invention except for the substrate being n-doped silicon. It would have been obvious to one having ordinary skill in the art at the time the invention was made to use n-doped silicon, since it has been held to be within the general skill of a worker in the art to select a known material on the basis of its suitability for the intended use as a matter of obvious design choice. In re Leshin, 125 USPQ 416.

BLS 3/22/04 Furthermore n-doped silicon will have higher electron mobility than silicon delivering better performance.

3. Claims 31,33-34, 36-37 and 39 are rejected under 35 U.S.C. 103(a) as be the teachings of Haukka et al. and Chon et al. unpatentable over Haukka et al. (US Pregrant Publication 2002/01 15252) in view of Chon et al. Haukka et al. disclose a gate dielectric with a substrate comprising of a semiconductor material , a metal oxide layer comprising an electrically insulating rare earth metal oxide disposed on the surface of the substrate , a conductive material on metal oxide layer and the conductive material acting as a gate electrode. However Haukka et al. fails to disclose an active element that changes with respect to applied voltage, resulting in a current voltage profile, and the charging and discharging of the metal oxide to form the current voltage profile (hysterisis curve). Whereas Chon et al. disclose a rare earth based memory element that has a hysterisis curve (active element that changes with respect to applied voltage,

resulting in a current voltage profile, and the charging and discharging of the metal oxide layer) (see figrue 2). Therefore it would have been obvious to one of ordinary skill in the ad at the time the invention was made to combine the teachings of Haukka et al. and Chon et al., because the use of hysterisis curves in memory applications is very well known in the art.

Response to Arguments

3. Applicant's arguments filed 12/31/03 have been fully considered but they are not persuasive.

4. In response to applicant's argument that claims 24 and 25 do have a predetermined current voltage-profile under an applied voltage and which forms an active element, a recitation of the intended use of the claimed invention must result in a structural difference between the claimed invention and the prior art in order to patentably distinguish the claimed invention from the prior art. If the prior art structure is capable of performing the intended use, then it meets the claim. See *In re Casey*, 152 USPQ 235 (CCPA 1967) and *In re Otto*, 136 USPQ 458, 459 (CCPA 1963).

5. In response to applicant's argument that the examiner's conclusion of obviousness is based upon improper hindsight reasoning, it must be recognized that any judgment on obviousness is in a sense necessarily a reconstruction based upon hindsight reasoning. But so long as it takes into account only knowledge which was within the level of ordinary skill at the time the claimed invention was made, and does not include knowledge gleaned only from the applicant's disclosure, such a reconstruction is proper. See *In re McLaughlin*, 443 F.2d 1392, 170 USPQ 209 (CCPA

1971). Furthermore, in response to applicant's argument that there is no suggestion to combine the references, the examiner recognizes that obviousness can only be established by combining or modifying the teachings of the prior art to produce the claimed invention where there is some teaching, suggestion, or motivation to do so found either in the references themselves or in the knowledge generally available to one of ordinary skill in the art. See *In re Fine*, 837 F.2d 1071, 5 USPQ2d 1596 (Fed. Cir. 1988)and *In re Jones*, 958 F.2d 347, 21 USPQ2d 1941 (Fed. Cir. 1992). In this case, Chon et al. disclose a capacitor that could be used in non-volatile memories (see abstract), and Huakka disclose the use of high dielectrics in gate electrodes, and furthermore high a high k dielectric structure in capacitor in integrated circuits (ie non-volatile memory)(see paragraph 0008).

6. In response to applicant's argument that Huakka fails to disclose a predetermined current voltage-profile under an applied voltage and which forms an active element, a recitation of the intended use of the claimed invention must result in a structural difference between the claimed invention and the prior art in order to patentably distinguish the claimed invention from the prior art. If the prior art structure is capable of performing the intended use, then it meets the claim. See *In re Casey*, 152 USPQ 235 (CCPA 1967) and *In re Otto*, 136 USPQ 458, 459 (CCPA 1963).

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Bradley K Smith whose telephone number is (571)272-1884. The examiner can normally be reached on 10-6 Monday through Friday.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Richard Elms can be reached on (571) 272-1869. The fax phone number for the organization where this application or proceeding is assigned is 703-872-9306.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

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Fatigue-free samarium-modified bismuth titanate ($Bi_{4-x}Sm_xTi_3O_{12}$) film capacitors having large spontaneous polarizations

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Fatigue-free and highly c-axis oriented $Bi_{3.15}Sm_{0.85}Ti_3O_{12}$ (BSmT) thin films were grown on $Pt/TiO_2/SiO_2/Si(100)$ substrates using the method of metalorganic sol decomposition. The BSmT film capacitor with a top Pt electrode showed significantly improved values of the remanent polarization ($2P_r$) and the nonvolatile charge as compared to those of the $Bi_{4-x}La_xTi_3O_{12}$ ($x=0.75$) film capacitor, recently known as the most promising candidate for nonvolatile memories. The $2P_r$ value of the BSmT capacitor was 49 $\mu C/cm^2$ at an applied voltage of 10 V while the net nonvolatile switching charge was as high as 20 $\mu C/cm^2$ and remained essentially constant up to 4.5×10^{10} read/write switching cycles at a frequency of 1 MHz. In addition to these, the capacitor demonstrated excellent charge-retention characteristics with its sensing margin of 17 $\mu C/cm^2$ and a strong resistance against the imprinting failure.
[DOI: 10.1063/1.1415353]

There have been extensive research efforts to enhance the reliability of perovskite-based ferroelectric thin films for use in nonvolatile ferroelectric random access memory (NvFRAM) devices. Among these ferroelectrics, lead zirconate titanate (PZT) is known to be the most important candidate for NvFRAM applications. However, it shows a serious degradation of ferroelectric properties after being subjected to $\sim10^7$ read/write switching cycles. Although the fatigue problem of PZT-based capacitors can be solved by using metal oxide electrodes,[1,2] these electrodes are difficult to be prepared and tend to increase leakage current, in general. Some layered perovskites such as strontium bismuth tantalate [$SrBi_2Ta_2O_9$, (SBT)][3,4] and lanthanum-modified bismuth titanate [$Bi_{3.25}La_{0.75}Ti_3O_{12}$, (BLT)][5] showed superior fatigue resistances as compared to Pt/PZT/Pt capacitors.

As one of the fatigue-free ferroelectrics, the BLT film is of particular interest. This is not only because it can be crystallized at low processing temperatures possibly below 650 °C, which is compatible with Si-based integrated circuit technology, but also because it shows larger spontaneous polarizations than those of SBT-based films. However, the BLT film prepared by the pulsed laser deposition method was characterized by a mixed orientation of grains.[5,6] The mixed orientation tends to increase bit-to-bit variability in a capacitor for high-density ferroelectric memory devices.[6] More recently, highly c-axis oriented BLT films having fatigue-free characteristics were grown on $Pt/Ti/SiO_2/Si(100)$ substrates using metalorganic sol decomposition (MOSD).[7] The highly c-axis oriented capacitor showed a well-saturated polarization–electric field ($P-E$) switching curve with its remanent polarization ($2P_r$) of 27 $\mu C/cm^2$ at an applied voltage of 10 V.[7] Notwithstanding the fatigue-free characteristics, the $2P_r$ value or, more relevantly, the nonvolatile switching charge of the highly oriented BLT film needs to be substantially improved to ensure the reliability of devices (i.e., sufficient sensing margin) and to apply the capacitor to high-density NvFRAM.

Our preliminary study indicated that the direction and the magnitude of $2P_r$ of highly c-axis oriented bismuth titanate (BT)-based films were very susceptible to the substitution of trivalent rare-earth lanthanides (e.g., La) for bismuth. X-ray diffraction (XRD) analysis of highly oriented BT-based films, in conjunction with ferroelectric measurements, indicated that the observed large increase in $2P_r$ values parallel to the c-axis with increasing content of lanthanides was closely related to the change in the major direction of the spontaneous polarization from the b-axis direction to the direction parallel to the c axis.[8] Thus, one would expect to obtain BT-based thin films having both fatigue-resistance characteristics and large $2P_r$ values by suitably substituting stable trivalent ions for volatile bismuth ions.

In selecting appropriate trivalent cationic species for this purpose, one has to consider various physicochemical factors. Among these, the following three factors seem to be most appropriate: (i) the stability of perovskite phase, (ii) the ionic radius, and (iii) the Curie–Weiss temperature. Among rare-earth lanthanides having electrons in the $4f$ orbitals, Sm, Nd, and Pr basically meet the requirement of the phase stability of $(BiO)_2R_2Ti_3O_{10}$-type layered perovskites.[9] Of these three lanthanides, samarium (Sm) seems to satisfy all of the three criteria. Its ionic radius of 1.00 Å in the Ahren's scale is compatible with that of Bi (0.93 Å).[9] The Curie–Weiss temperature of $Bi_{3.15}Sm_{0.85}Ti_3O_{12}$ is estimated to be 470 °C,[9] which is high enough to be used in ferroelectric memory devices that require a stability of polarization switching against thermal agitation. In view of these, the

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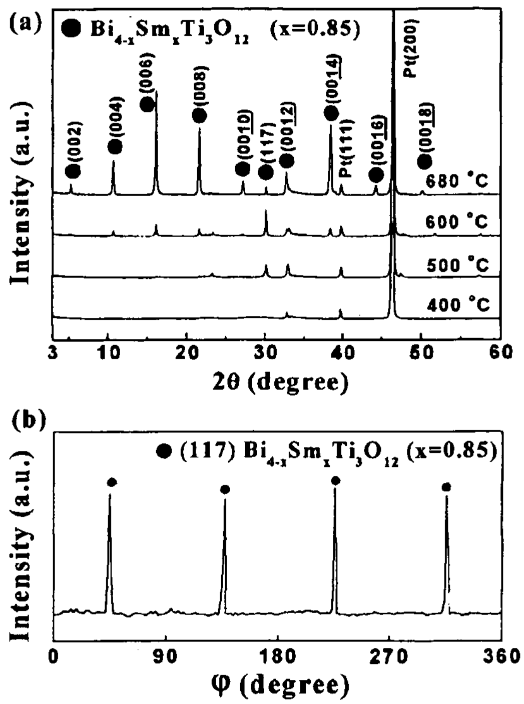

FIG. 1. XRD θ–2θ patterns and φ-scan spectrum of BSmT films grown on $Pt/TiO_2/SiO_2/Si(100)$ substrates (a) XRD θ–2θ scan results of BSmT films annealed at various indicated temperatures for 1 h, and (b) φ-scan spectrum for the (117) reflection of the c-axis oriented BSmT film annealed at 680 °C for 1 h.

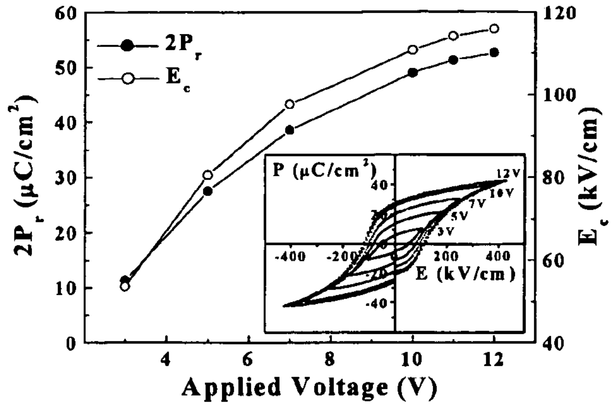

FIG. 2. Variations of $2P_r$ and E_c values of the Pt/BSmT/Pt capacitor are plotted as a function of the applied voltage. The inset shows hysteresis loops measured at various applied voltages ranging between 3 and 12 V. The corresponding film was thermally annealed at 680 °C for 1h.

main purpose of the present study is to develop highly c-axis oriented Sm-modified bismuth titanate (BSmT) films having fatigue-free characteristics as well as improved remananent polarizations along the unique c direction.

The BSmT ($Bi_{4-x}Sm_xTi_3O_{12}$ with $x=0.85$) films were fabricated on $Pt/TiO_2/SiO_2/Si(100)$ substrates using the method of MOSD. The precursor sol for the coating was prepared by dissolving appropriate amounts of bismuth acetate $[Bi(CH_3COO)_3]$, samarium acetate hydrate $[Sm(CH_3COO)_3 \cdot 2H_2O]$, and titanium isopropoxide $\{Ti[(CH_3)_2CHO]_4\}$ in an acetic acid solution at room temperature in a glove box being flushed with nitrogen gas. The dried amorphous films were crystallized by thermal annealing in an oxygen-rich atmosphere at various temperatures ranging between 400 °C and 680 °C for 1 h.

As-annealed films were specular, crack-free, dense, and adhered well on the substrates used. Microstructural examination using a field-emission scanning electron microscope (FE-SEM) showed only fine-sized, uniform grains in the films. The chemical composition of the BSmT film, as determined using energy dispersive x-ray and electron microprobe techniques, was Bi:Sm:Ti=3.15:0.85:3. In order to fabricate capacitors, top Pt electrodes were deposited using a rf magnetron sputter. A typical area of the top electrode was 10^{-4} cm^2. The ferroelectric and dielectric measurements were performed on the BSmT capacitors using a RT6000S ferroelectric tester and a HP4194A impedance analyzer equipped with a micrometer probe station.

Figure 1 shows the XRD θ–2θ scan results of the BSmT films annealed at various indicated temperatures for 1 h. All the XRD patterns can be readily identified and indexed using the standard XRD data for the perovskite BT ($Bi_4Ti_3O_{12}$) compiled in the Joint Committee on Powder Diffraction Standards card. This indicates that the BSmT film maintains a pseudotetragonal-layered structure similar to the perovskite BT even under extensive modifications by Sm. The most prominent feature of the XRD patterns in Fig. 1(a) is that the film annealed at 680 °C shows a highly c-axis oriented preferential growth with a minor fraction of (117) orientation. The degree of the $(00l)$-type preferential growth, as estimated using Lotgering's orientation factor,[10] is 96% for the BSmT film annealed at 680 °C. The x-ray φ-scan spectrum for the (117) reflection shows four peaks separated one from another by 90°, as presented in Fig. 1(b). This indicates that the c-axis oriented tetragonal BSmT film ($x=0.85$) has a homogeneous in-plane orientation.

Figure 2 summarizes the variations of $2P_r$ and E_c (coercive field) of the BSmT capacitor with the applied voltage. The film was annealed at 680 °C for 1 h, and its thickness, as estimated using cross sectional FE-SEM, was 280 nm. The inset presents hysteresis loops measured at various applied voltages ranging between 3 and 12 V. The capacitor is characterized by well-saturated $P-E$ switching curves. As shown in Fig. 2, both $2P_r$ and E_c values increase rather steeply at a low applied voltage but do not change much beyond 10 V. The capacitor is nearly saturated at an applied voltage of 12 V (i.e., ~430 kV/cm). $2P_r$ value of the capacitor is 49 μC/cm^2 at an applied voltage of 10 V. This value is remarkably higher than $2P_r$ of 27 μC/cm^2 for the highly c-axis oriented $Bi_{3.25}La_{0.75}Ti_3O_{12}$ capacitor, recently reported as a fatigue-free ferroelectric capacitor.[7]

The relative dielectric permittivity $[\varepsilon'(\omega)]$ and the dissipation factor $[\varepsilon''(\omega)/\varepsilon'(\omega)=\tan\delta]$ of the capacitor were measured at 25 °C as a function of frequency. $\varepsilon'(\omega)$ and $\tan\delta$ were 387 and 0.054 at a frequency of 1 MHz, respectively. These values are comparable to those of PZT, SBT, and BLT capacitors.[3,5,11] Although both $\varepsilon'(\omega)$ and $\tan\delta$ decreased slightly with increasing frequency, there was no sudden change in their values up to 1 MHz. All these indicate that the observed $P-E$ hysteresis behavior of the BSmT capacitor originates from the ferroelectric polarization switching of bound charges, not from the response of freely moving charges.

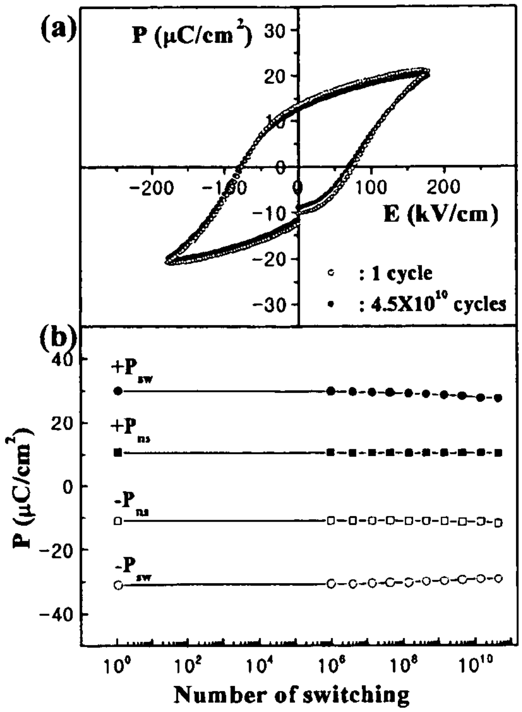

FIG. 3. Electrical fatigue characteristics of the Pt/BSmT/Pt capacitor are shown before and after being subjected to 4.5×10^{10} read/write cycles at a frequency of 1 MHz. (a) $P-E$ hysteresis loops measured at an applied voltage of 5 V before and after the switching cycle, and (b) the fatigue test results determined using a switching (fatigue) voltage of ±3 V, and a measuring voltage of 5 V at a frequency of 1 MHz.

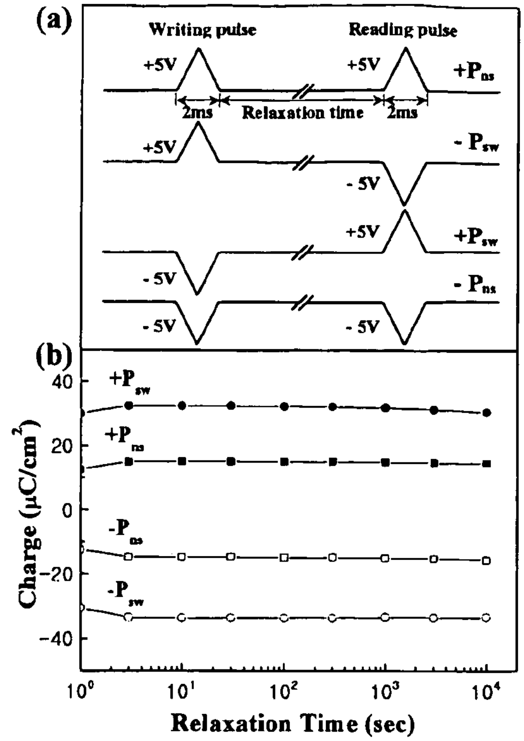

FIG. 4. Charge–retention characteristics of the Pt/BSmT/Pt capacitor at 85 °C are plotted as a function of time after the application of a writing pulse.

The fatigue-free characteristics of the BSmT capacitor are summarized in Fig. 3. The capacitor shows little change both in the switching polarization (P_{sw}) and in the nonswitching polarization (P_{ns}) up to 4.5×10^{10} read/write cycles at a switching voltage of ±3 V (i.e., 107 kV/cm). The values of the nonvolatile charge [i.e., $(+P_{sw})-(+P_{ns})$ or $(-P_{sw})-(-P_{ns})$] are approximately 20 μC/cm^2, and remain essentially constant throughout the switching cycles. The fatigue-free behavior was also observed at a higher switching voltage of ±5 V (i.e., 180 kV/cm). The $P-E$ curves of Fig. 3(a) were obtained at an applied voltage of 5 V before and after the electrical fatigue test. The values of $2P_r$ and E_c before the fatigue test were 27 μC/cm^2 and 79 kV/cm, respectively. After being subjected to 4.5×10^{10} cycles, these were still retained at 25 μC/cm^2 and 76 kV/cm. Besides, the $P-E$ curves do not show any noticeable asymmetric behavior resulting in imprint failures, even after being subjected to 4.5×10^{10} switching cycles.

The charge–retention characteristics of the BSmT capacitor are summarized in Fig. 4 by plotting the switching polarization ($\pm P_{sw}$) and the nonswitching polarization ($\pm P_{ns}$) as a function of time. Figure 4(a) represents four distinctive test-pulse sequences employed for measuring the charge retention of $+P_{sw}$, $+P_{ns}$, $-P_{sw}$, and $-P_{ns}$. The sensing margin, as defined by $P_{nv}=(\pm P_{sw})-(\pm P_{ns})$, of the capacitor was 17 μC/cm^2 at 85 °C and remained essentially constant up to 10^4 s after applying a writing pulse, demonstrating an excellent charge-retaining ability of the BSmT capacitor. Compared with the reported sensing margin of a highly c-axis oriented SBT capacitor[12] (6 μC/cm^2) and that of a $SrBi_2(Ta,Nb)_2O_9$ capacitor[13] (11 μC/cm^2), this is a substantial improvement. In addition to this, $|-P_{sw}|$ was essentially the same as $+P_{sw}$ throughout the relaxation (retaining) time, and the same trend was also observed for $|-P_{ns}|$ and $+P_{ns}$. This indicates that the BSmT capacitor has a strong resistance against the imprinting failure.

This study was financially supported by the KISTEP of Korea through the NRL Program.

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